

STUDY OF THE RADIOACTIVITY INDUCED IN AIR BY A 15-MeV PROTON BEAM

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Radioactivity induced by a 15-MeV proton beam extracted into air was studied at the beam transport line of the 18-MeV cyclotron at the Bern University Hospital (Inselspital). The produced radioactivity was calculated and measured by means of proportional counters located at the main exhaust of the laboratory. These devices were designed for precise assessment of air contamination for radiation protection purposes. The main produced isotopes were ¹¹C, ¹³N and ¹⁴O. Both measurements and calculations correspond to two different irradiation conditions. In the former, protons were allowed to travel for their full range in air. In the latter, they were stopped at the distance of 1.5 m by a beam dump. Radioactivity was measured continuously in the exhausted air starting from 2 min after the end of irradiation. For this reason, the short-lived ¹⁴O isotope gave a negligible contribution to the measured activity. Good agreement was found between the measurements and the calculations within the estimated uncertainties. Currents in the range of 120–370 nA were extracted in air for 10–30 s producing activities of 9–22 MBq of ¹¹C and ¹³N. The total activities for ¹¹C and ¹³N per beam current and irradiation time for the former and the latter irradiation conditions were measured to be $(3.60 \pm 0.48) \times 10^{-3}$ MBq (nA s)⁻¹ and $(2.89 \pm 0.37) \times 10^{-3}$ MBq (nA s)⁻¹, respectively.

INTRODUCTION

The assessment of the radioactivity present in air is an important issue in radiation protection. A reliable and efficient monitoring is crucial for radioisotope production and research facilities. Typical examples are positron emission tomography (PET) radioisotope production centres, where ⁴¹Ar is regularly produced in air during irradiation through the reaction ⁴⁰Ar(n,γ)⁴¹Ar. Furthermore, releases of β⁺ emitters in gas form (mainly ¹³N and ¹⁸F) may provoke radiological hazards.

Some research applications require the extraction of the beam into air as, for example, PIXE and PIGE ion beam analysis and the bombardment of cell samples for radiation biology. For these kinds of experimental activities, radioisotopes are produced in air by nuclear reactions induced by the beam and an accurate assessment of the radioactivity is therefore needed.

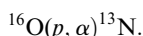
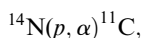
In this work, a study of the radioactivity produced by a 15-MeV proton beam extracted into air is presented. This work was performed at the cyclotron laboratory at the Bern University Hospital (Inselspital), which was realised for PET radioisotope production and multi-disciplinary research running in parallel^(1, 2). This facility is equipped with a beam transport line ending in a separate bunker⁽³⁾, where the experiments reported in this study were performed. Specific detectors assure the continuous radiation protection monitoring of exhausted air. Low-sensitivity alarm

detectors are located at the exhaust of each bunker and high-sensitivity proportional counters⁽⁴⁾ are placed at the main exhaust of the facility to assess the integral of the activity released into the atmosphere. These proportional counters were used to measure the radioactivity produced by extracting beams of known current into air. Calculations of the induced activity were performed and compared with the data.

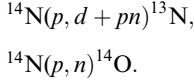
CALCULATION OF INDUCED RADIOACTIVITY

When a 15-MeV proton beam is extracted into air, radioactive isotope production occurs. The nuclei present in air can undergo nuclear reactions. Air is composed of nitrogen, oxygen, argon, carbon and traces of other elements. Nitrogen (78.084 %) and oxygen (20.948 %) represent by far the main components⁽⁵⁾. Concerning their isotope abundance, ¹⁴N accounts for ~99.5 % and ¹⁶O for ~99.8 %.

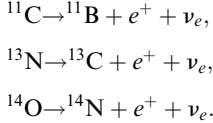
To assess the nuclear reactions, cross sections for the production of various isotopes by protons in air⁽⁶⁾ together with target concentrations were considered. Two reactions were found to be dominant:



Two further reactions involving ^{14}N as a target were also considered due to their non-negligible cross section:



Produced isotopes decay according to the following reactions:



Since the reactions are three-body decays, the maximum energies of the positron for ^{11}C and ^{13}N are 0.96 and 1.19 MeV, respectively. For ^{14}O , the maximum positron energy is 1.81 MeV in 99.4 % of the decays and 4.12 MeV in the remaining 0.6 %.

The production rates of ^{11}C , ^{13}N and ^{14}O are indicated as P_C , P_N and P_O , respectively. The production rate P (in nuclei s^{-1}) for one reaction is given by the formula:

$$P = \frac{n \cdot I}{e} \int_{E_i}^{E_f} \frac{\sigma(E)}{dE/dx} dE, \quad (1)$$

where n is the target density (in nuclei m^{-3}), I the beam current, e the elementary charge, σ the reaction cross section, E the energy of the incident particle and x the travelled distance. The integral was calculated from the initial energy E_i of the proton to its final energy E_f . Since protons are accelerated by the Bern cyclotron to the energy of 18 MeV and passed through a 400- μm thick aluminium exit window, the initial energy E_i of protons extracted into air was calculated by means of the simulation software SRIM⁽⁷⁾. E_i was found to be 15.56 ± 0.10 MeV. In the case of the full proton range in air, E_f is equal to 0. If the beam dump at 1.5 m from the exit window was used, E_f was also determined by SRIM. It was found to be 9.55 ± 0.10 MeV. The reaction cross sections $\sigma(E)$ were evaluated on the basis of the measurements reported in refs^(6, 8, 9). The energy loss per unit length dE/dx used for the evaluation of the integral was calculated by means of SRIM. Dry air near the sea level (International Commission on Radiation Units and Measurements-104) was considered. On this basis, the integral was computed numerically.

The resulting value P was then used for the evaluation of the corresponding produced radioactivity. The number of radioactive nuclei N present in air is a function of the irradiation time t and is derived from

the following equation:

$$\frac{dN(t)}{dt} = -\lambda N(t) + P, \quad (2)$$

where λ is the decay constant of the particular isotope: $\lambda_C = 5.669 \times 10^{-4} \text{ s}^{-1}$, $\lambda_N = 1.159 \times 10^{-3} \text{ s}^{-1}$ and $\lambda_O = 9.818 \times 10^{-3} \text{ s}^{-1}$ for ^{11}C , ^{13}N and ^{14}O , respectively. This leads to the expression for the activity A at time t :

$$A(t) = P(1 - e^{-\lambda t}). \quad (3)$$

The results of the calculations for the full proton range in air as well as for protons stopped at the distance of 1.5 m are presented in Table 1. For ^{13}N , the result is obtained by summing the rates given by the two production reactions. The production of ^{11}C was found to be dominant. The uncertainties of the performed calculations are due to the measured excitation functions^(6, 8, 9).

As shown in Figure 1, the contribution to the total activity due to ^{14}O rapidly decreases in time due to its

Table 1. Production rates per beam current are reported for full proton range and protons stopped at the distance of 1.5 m.

Full proton range	
$P_C/I [\text{nuclei (nA s)}^{-1}]$	$(5.31 \pm 1.14) \times 10^6$
$P_N/I [\text{nuclei (nA s)}^{-1}]$	$(8.15 \pm 1.28) \times 10^5$
$P_O/I [\text{nuclei (nA s)}^{-1}]$	$(2.40 \pm 1.16) \times 10^5$
Protons stopped at 1.5 m	
$P_C/I [\text{nuclei (nA s)}^{-1}]$	$(3.52 \pm 0.66) \times 10^6$
$P_N/I [\text{nuclei (nA s)}^{-1}]$	$(7.56 \pm 1.01) \times 10^5$
$P_O/I [\text{nuclei (nA s)}^{-1}]$	$(1.72 \pm 0.82) \times 10^5$

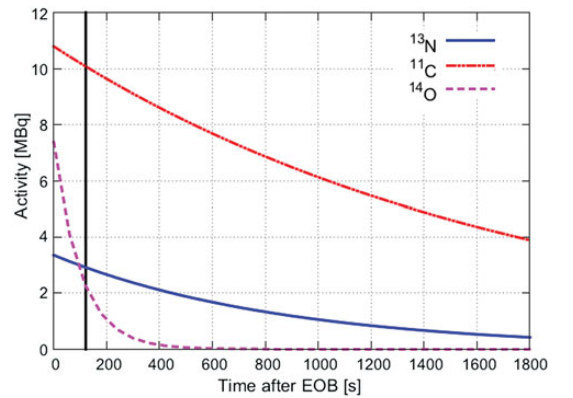


Figure 1. Time evolution of the total calculated activity for ^{11}C , ^{13}N and ^{14}O in the full proton range irradiation condition. The EOB is represented by $t = 0$. The vertical line corresponds to the start of the measurements, 2 min after EOB. A very similar plot was obtained for the other irradiation condition.

much shorter lifetime. For this reason, the measuring method used for this study is almost insensitive to ^{14}O .

EXPERIMENTAL SET-UP

The external beam transport line (BTL) of the Bern cyclotron was used for the measurements⁽³⁾. The cyclotron provides 18-MeV proton beams of variable intensity up to 150 μA . The BTL is equipped with an up-stream collimator, X-Y steering magnets, two quadrupole doublets, a neutron shutter, two beam viewers with Faraday cups for measuring the current and a beam dump. The beam line ends in a separate bunker. A schematic view of the cyclotron and the BTL is presented in Figure 2. For the measurements reported in this study, the beam dump was replaced by a 400- μm thick aluminium exit window.

The laboratory is equipped with high-sensitivity detectors for the continuous measurement of radioactivity in exhausted air⁽⁴⁾. These detectors are installed inside the main exhaust pipe of the facility, a few metres before the chimney. They consist of two large surface proportional counters with an active area of 1000 cm^2 . The detectors are filled with argon-methane for low background counting rate. They are

calibrated for ^{18}F , which is daily produced for the synthesis of PET radiotracers. Since the decays of ^{11}C and ^{13}N were considered in this experiment, the scaling factor f given by the manufacturer⁽⁴⁾ was applied. It was determined to be 0.6 for ^{13}N . Although the maximum positron decay energies for these two radionuclides are slightly different, a single scaling factor was considered. This represents a good approximation for this study and was taken into account in the estimation of the uncertainty. The air monitoring system provides diverse estimators of the exhausted air contamination. In this experiment, the so-called chimney short emission value (CSEV) was used. It is computed on the basis of the measured activity per volume (in kBq m^{-3}) and of the air flow (in $\text{m}^3 \text{h}^{-1}$). The CSEV is expressed in kBq h^{-1} and is updated every minute. The ventilation system runs continuously and was set at 5.5 air changes per hour in the BTL bunker (volume 63 m^3). For the whole facility, $\sim 20\,000 \text{ m}^3 \text{h}^{-1}$ of air was exhausted to the atmosphere. Due to the transit time of the air in the ventilation system, radioactivity produced in the air of the BTL bunker was measured with a delay of ~ 2 min.

For the full proton range measurement, the intensity of extracted current was measured with a movable stopper made of aluminium. The movement of the

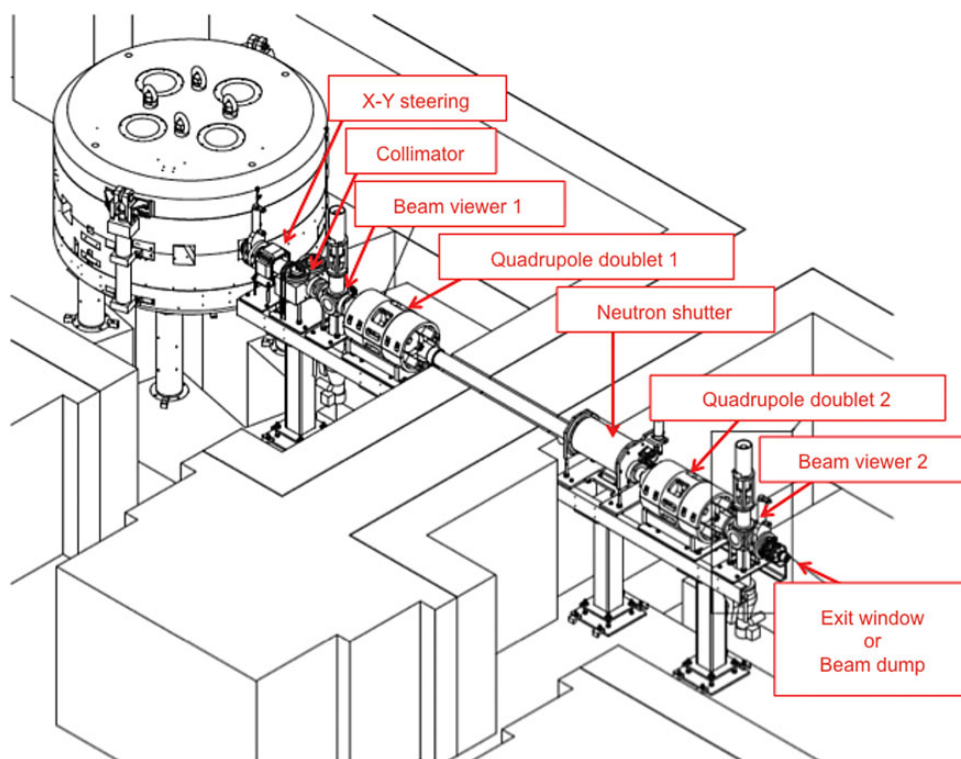


Figure 2. Schematic view of the Bern cyclotron and its beam transport line.

stopper was automatised so that the beam could be extracted into air right after the measurement of the intensity. This system is shown in Figure 3. In this way, protons were extracted into air and fully stopped inside the bunker. The range of a 15-MeV proton

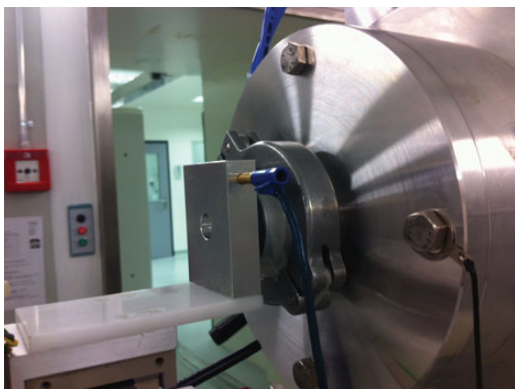


Figure 3. Motorised movable stopper for beam current measurements.

beam in dry air calculated by means of SRIM was found to be 2.60 m. The distance between the aluminium exit window and the bunker wall is >3 m so that the beam is fully stopped in air. In the case of the measurement with the beam dump, protons travelled in air at a distance of 1.50 m and were stopped by a 20-mm thick aluminium plate. This configuration is shown in Figure 4. The current measured on the beam dump corresponds to the beam intensity, since the fraction of protons undergoing nuclear reactions or large angle scattering effects is negligible. Each beam extraction in both irradiation conditions was followed by ~ 45 min of data taking with the air monitoring system. The next irradiation was performed only when the activity reached again the level of the background, as presented in Figure 5.

DATA ANALYSIS

Data recorded by the high-sensitivity proportional counters installed in the main exhaust allowed estimating the radioactivity produced during the irradiation. As shown in Figure 1, the contribution due to ^{14}O is almost negligible and was considered only for

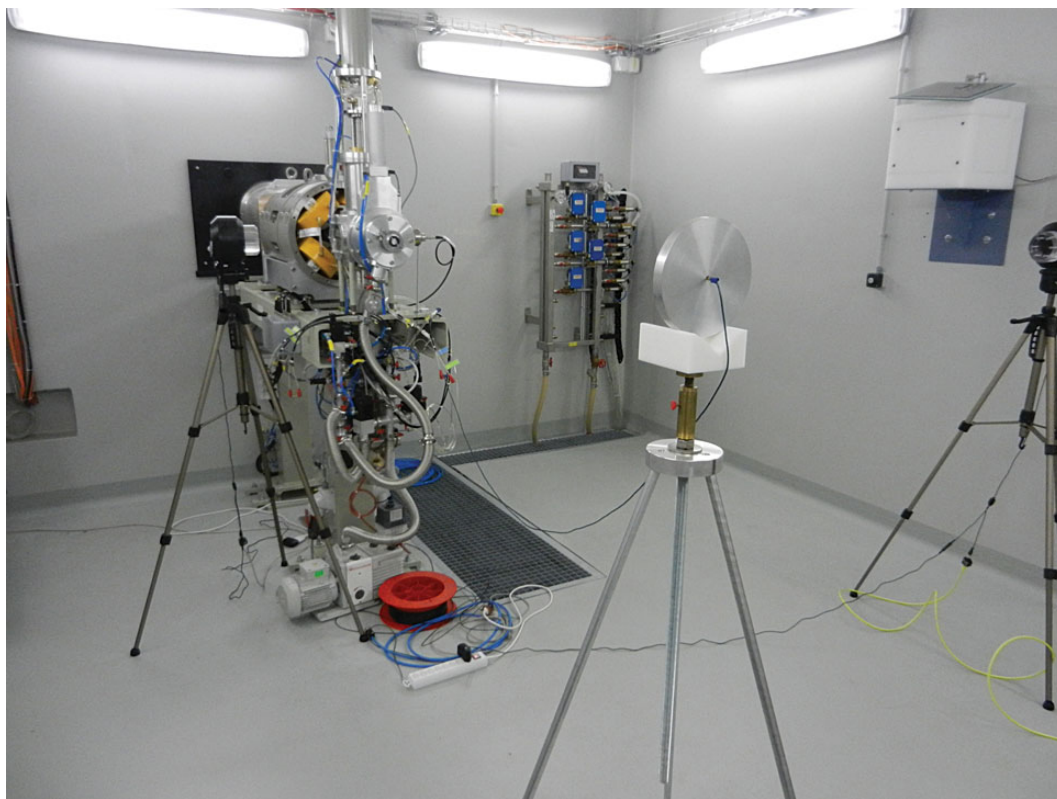


Figure 4. Set-up for the measurement with a beam dump at the distance of 1.5 m.

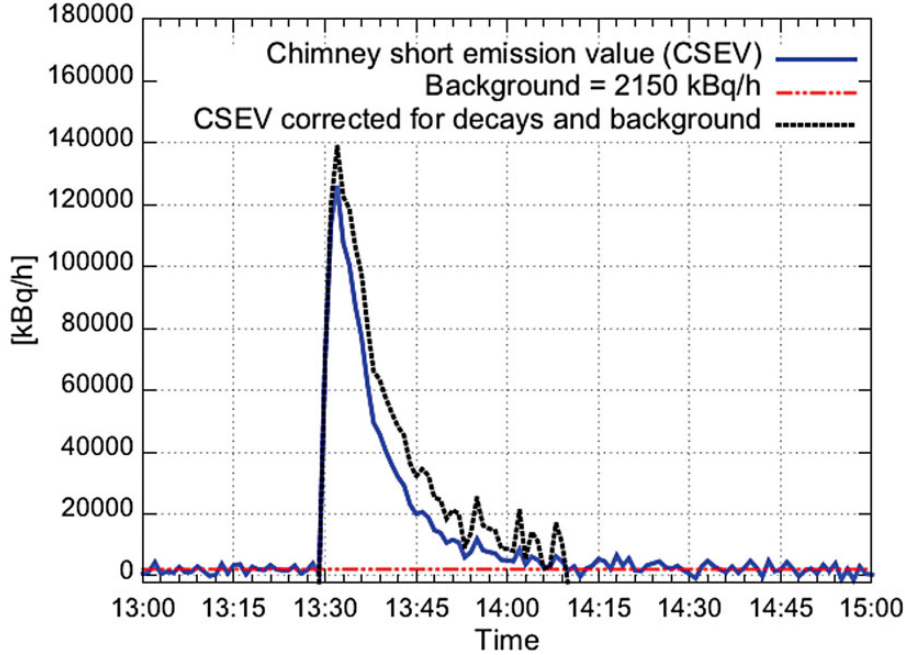


Figure 5. The CSEVs as a function of time for one irradiation (solid line) and the corresponding estimated background (dashed-dotted line). To calculate the produced activities at EOB $A_i(t_0)$, the CSEVs were corrected for decay and background (dotted line). The EOB happened 2 min before the rise of the signal.

the estimation of the uncertainty. Since no other experimental activities were carried out in the whole facility during the measurements, no contamination due to other isotopes was possible. The activities A_C and A_N —corresponding to ^{11}C and ^{13}N , respectively—are therefore considered for the data analysis. Considering the decay constants λ_C and λ_N , the time evolution of A_C and A_N is given by

$$A_C(t) = A_C(t_0)e^{-\lambda_C t}, \quad (4)$$

$$A_N(t) = A_N(t_0)e^{-\lambda_N t}, \quad (5)$$

where t_0 corresponds to the end of bombardment (EOB). The total induced activity at EOB $A_{\text{tot}}(t_0)$ is given by

$$A_{\text{tot}}(t_0) = A_C(t_0) + A_N(t_0), \quad (6)$$

and was obtained through the measurements of the CSEVs performed at successive time instants t_i , as presented in Figure 5 for one of the irradiations. Due to the continuous air flow in the ventilation system, signals different from background are recorded up to ~ 40 min after EOB.

Since the total activity was measured, the contributions of ^{11}C and ^{13}N were estimated on the basis of

the calculated values $A_C^{\text{calc}}(t_0)$ and $A_N^{\text{calc}}(t_0)$. The factor k_N is introduced and represents the fraction of the total activity due to ^{13}N at t_0 :

$$k_N = \frac{A_N^{\text{calc}}(t_0)}{A_C^{\text{calc}}(t_0) + A_N^{\text{calc}}(t_0)}. \quad (7)$$

The factor k_N is equal to 0.24 and 0.31 for the full proton range and for protons stopped at 1.5 m, respectively. Small variations of k_N due to the irradiation time were taken into account. This leads to the formula for the activity $A_i(t_i)$ corresponding to the i th measurement of the radioactivity released in air:

$$A_i(t_i) = A_i(t_0)(k_N e^{-\lambda_N t_i} + (1 - k_N) e^{-\lambda_C t_i}). \quad (8)$$

The total produced activity $A_{\text{tot}}(t_0)$ is the sum of all the activities $A_i(t_0)$:

$$A_{\text{tot}}(t_0) = \sum_i A_i(t_0) \quad (9)$$

$$= f \sum_i \frac{\epsilon_i}{k_N e^{-\lambda_N t_i} + (1 - k_N) e^{-\lambda_C t_i}}, \quad (10)$$

where ϵ_i is the CSEV multiplied by the time interval

Table 2. Summary of the measurements performed in the two irradiation conditions together with the corresponding calculated values.

#	Beam current [nA]	Irradiation time [s]	$A_{\text{tot}}(t_0)$ [MBq]	A^{calc} [MBq]
Full proton range				
1	120	30	12.28 ± 0.88	14.15 ± 2.37
2	260	15	13.74 ± 0.95	15.35 ± 2.57
3	280	20	22.66 ± 1.50	22.00 ± 3.69
4	260	20	17.78 ± 1.11	20.43 ± 3.42
Protons stopped at 1.5 m				
1	370	10	10.37 ± 0.76	10.44 ± 1.43
2	170	20	10.35 ± 0.77	9.55 ± 1.31
3	220	30	19.11 ± 1.27	18.48 ± 2.53
4	260	30	22.07 ± 1.48	21.84 ± 2.99

A^{calc} corresponds to the contributions of ^{11}C and ^{13}N .

Table 3. Experimental results compared with calculations. Total activities per beam current and irradiation time are reported for full proton range and protons stopped at the distance of 1.5 m. A^{calc} corresponds to the contributions of ^{11}C and ^{13}N .

$A_{\text{tot}}(t_0)/(I \cdot t)$ [MBq (nA s) $^{-1}$]	$A^{\text{calc}}/(I \cdot t)$ [MBq (nA s) $^{-1}$]
Full proton range $(3.60 \pm 0.48) \times 10^{-3}$	$(3.93 \pm 0.66) \times 10^{-3}$
Protons stopped at 1.5 m $(2.89 \pm 0.37) \times 10^{-3}$	$(2.81 \pm 0.39) \times 10^{-3}$

of one minute and $f = 0.6$ is the scaling factor of the proportional counters for ^{11}C and ^{13}N .

For the two different irradiation conditions, four measurements were performed. They are reported in Table 2 together with the calculated values A^{calc} , which include the contributions of ^{11}C and ^{13}N . Good agreement is found between measurements and calculations.

According to Equation (3), the produced activity $A(t)$ can be linearly approximated for $t \ll 1/\lambda$, where t denotes the irradiation time. Since this condition was fulfilled in the reported measurements, the average values of $A_{\text{tot}}(t_0)$ per beam current I and irradiation time t were calculated for both irradiation conditions.

The results compared with the corresponding values of $A^{\text{calc}}/(I \cdot t)$ are presented in Table 3.

The main source of uncertainty is the precision of the air monitoring system, which is estimated by the manufacturer to be 20 % of the measured value. Statistical uncertainties are significant and account for up to ~30 % of the total uncertainties. Other relevant sources of uncertainty are the beam current (≈ 10 %), the scaling factor of the proportional

counters (≈ 10 %), the irradiation time (≈ 5 %), the k_N factor (≈ 5 %) and the presence of a residual contribution due to ^{14}O (≈ 4 %).

At the cyclotron laboratory in Bern, the alarm threshold for air contamination is set at 70 kBq m $^{-3}$ at the main exhaust and, according to the Swiss regulations, a maximum of 784 MBq can be exhausted per week. If the alarm threshold is overcome, the ventilation of the whole facility shuts down, provoking severe consequences for the good manufacturing practice (GMP) radiopharmaceutical production and for radiation protection procedures. For these reasons, extreme care is needed when research activities requiring the use of the proton beam extracted into air are performed. As an example, the extraction of 500 nA for 60 s producing ~100 MBq of activity is sufficient to trigger the alarm.

CONCLUSIONS

The activity induced by a 15-MeV proton beam extracted into air was measured and calculated for two irradiation conditions. In the former, protons were allowed to travel for their full range in air. In the latter, the beam was stopped at the distance of 1.5 m by an aluminium beam dump.

Calculations were based on excitation functions available in the literature. The experimental work was performed with the beam transport line of the 18-MeV cyclotron at the Bern University Hospital (Inselspital). The produced radioactivity was measured by means of large surface proportional counters located at the main exhaust of the cyclotron laboratory. Proton beams in the current range of 120–370 nA were extracted into air for a duration of 10–30 s.

In both irradiation conditions, the main produced isotopes were ^{11}C , ^{13}N and ^{14}O . Due to the transit time of the air flow between the beam line bunker and the main exhaust of ~2 min, the contribution to the measurements given by the short-lived ^{14}O was negligible. The experimental results are found to be in good agreement with the calculations for ^{11}C and ^{13}N within the total uncertainties. For irradiation times much shorter than ^{11}C and ^{13}N lifetimes, the induced total activities per beam current and irradiation time were measured to be $(3.60 \pm 0.48) \times 10^{-3}$ MBq (nA s) $^{-1}$ and $(2.89 \pm 0.37) \times 10^{-3}$ MBq (nA s) $^{-1}$, for the former and for the latter irradiation conditions, respectively.

The extraction of proton beams into air requires particular care, especially if performed in PET radiopharmaceutical production centres. The produced activity may induce alarms, ventilation shut downs and radiation protection issues with direct consequences for the production. The results reported in this study can be used for the assessment of radioactivity induced in air by proton beams in the energy range of PET cyclotrons.

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REFERENCES

1. Braccini, S. *et al.* SWAN: a combined centre for radioisotope production, proton therapy and research in Bern. In: Presented at the Workshop on Physics for Health in Europe, CERN, Geneva, Switzerland (2010).
2. Braccini, S. *et al.* The new Bern cyclotron laboratory for radioisotope production and research. In: Proceedings of the Second International Particle Accelerator Conference (IPAC2011), San Sebastian, Spain, 3618 (2011).
3. Braccini, S. The new Bern PET cyclotron, its research beam line, and the development of an innovative beam monitor detector. In: Proceedings of the 22nd International Conference on the Application of Accelerators in Research and Industry (CAARI 2012), Forth Worth, Texas, USA (2012).
4. Berthold Technologies GmbH. Radiation protection for PET isotope centers (2008).
5. (2003) *CRC Handbook of Chemistry and Physics*, eighty-fourth edn, CRC Press.
6. International Atomic Energy Agency. *Experimental nuclear reaction data (EXFOR)*. IAEA. <http://www-pub.iaea.org>. (accessed 19 June, 2014).
7. Ziegler, J. F. and Manoyan, J. M. The stopping of ions in compounds. Nucl. Instrum. Methods B **35**, 215, SRIM-2013.00 (1988). <http://www.srim.org>. (accessed 19 June, 2014).
8. International Atomic Energy Agency. *Technical Reports Series No. 468: Cyclotron produced radionuclides: physical characteristics and production methods* (2009).
9. Kovács, Z., *et al.* Cross section measurements using gas and solid targets for production of the positron-emitting radionuclide O-14. Radiochim. Acta **91**, 185 (2003).